

COOL-COLOR ROOFING MATERIAL ATTACHMENT 5: TASK 2.5.2 REPORTS -DESIGN INNOVATIVE METHODS FOR APPLICATION OF COOL COATINGS TO ROOFING MATERIALS

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Methods of Creating Solar-Reflective Nonwhite Surfaces and their Application to Residential Roofing Materials

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Abstract

We describe methods for creating solar-reflective nonwhite surfaces and their application to a wide variety of residential roofing materials, including metal, clay tile, concrete tile, wood, and asphalt shingle. Reflectance in the near-infrared (NIR) spectrum $(0.7 - 2.5 \mu m)$ is maximized by

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coloring a topcoat with pigments that weakly absorb and (optionally) strongly backscatter NIR radiation and adding an NIR-reflective basecoat (e.g., titanium dioxide white) if both the topcoat and the substrate weakly reflect NIR radiation. Coated steel and glazed clay tile roofing products achieved NIR reflectances of up to 0.50 and 0.75, respectively, using only cool topcoats. Gray concrete tiles achieved NIR reflectances as high as 0.60 with coatings colored by NIR-scattering pigments. Such tiles could attain NIR reflectances of up to 0.85 by overlaying a white basecoat with a topcoat colored by NIR-transparent organic pigments. Granule-surfaced asphalt shingles achieved NIR reflectances as high as 0.45 when the granules were covered with a white basecoat and a cool color topcoat.

Introduction

A roof with high solar reflectance (ability to reflect sunlight) and high thermal emittance (ability to radiate heat) stays cool in the sun, reducing demand for cooling power in conditioned buildings and increasing occupant comfort in unconditioned buildings. Nonmetallic surfaces and most polymer-coated metal surfaces have high thermal emittance. Hence, a cool roofing surface may be described as a nonmetal or polymer-coated metal with high solar reflectance.

Visible light $(0.4-0.7~\mu m)^{-1}$ contains 43% of the power in the air-mass 1.5 global solar irradiance spectrum $(0.3-2.5~\mu m)$ typical of North-American ground-level insolation; the remainder arrives as near-infrared (NIR) radiation $(0.7-2.5~\mu m, 52\%)$ or ultraviolet (UV) radiation $(0.3-0.4~\mu m, 5\%)$ (ASTM 2003). A clean, smooth, and solar-opaque white surface strongly reflects both visible and NIR radiation, achieving a solar reflectance of about 0.85. This is the coolest type of roofing surface, and is ideal for low-slope roofs visible neither from ground level nor from taller buildings.

The solar reflectance of a roofing surface (especially that on a home) may be constrained by (a) desire for a nonwhite appearance, which limits visible reflectance; (b) NIR transparency of a thin and/or sparsely pigmented coating; and/or (c) curvature, which can cause light reflected from one face to be absorbed by another face. Nonwhite surfaces can be made as cool as possible by maximizing reflectance in the NIR spectrum, which does not affect color. Smoothing rough surfaces can increase reflectance at all wavelengths.

This study describes the engineering principles for creating a solar-reflective coated surface, and their application to a wide variety of residential roofing materials, including metal, clay tile, concrete tile, wood, and asphalt shingle.

Literature Review

Brady and Wake (1992) present the basic method for creating a coating with high NIR reflectance: color an otherwise transparent topcoat with pigments that weakly absorb and (optionally) strongly backscatter NIR radiation, adding an NIR-reflective basecoat (e.g., titanium

¹ The spectrum of visible light is typically specified as either 0.38 - 0.78 μm, or 0.40 - 0.70 μm. We choose the simpler range 0.40 to 0.70 μm because phototropic responses to light in the tails (0.38 - 0.40 μm and 0.70 - 0.78 μm) are low (ASTM 2001).

dioxide white) if both the topcoat and the substrate weakly reflect NIR radiation (Figure 1). This technique is reprised in whole or in part by U.S. patents and patent applications for creating generic NIR-reflectors (Genjima and Haruhiko 2002; Hugo 2002) and for creating NIR-reflective granules and/or granule-surfaced asphalt shingles (Gross and Graham 2005; Joedicke 2003; Ming et al. 2005a,b).

The authors reviewed current methods of manufacturing metal, clay tile, concrete tile, and asphalt shingle roofing materials in a earlier pair of articles (Akbari et al. 2005a,b).

Methodology

Maximizing solar reflectance of a colored surface

The fraction R of solar radiation incident at wavelengths between λ_0 and λ_1 that is reflected by a surface is the irradiance-weighted average of the surface's spectral reflectance $r(\lambda)$. That is,

$$R_{\lambda_0 \to \lambda_1} = \left(\int_{\lambda_0}^{\lambda_1} r(\lambda) i(\lambda) d\lambda \right) / \int_{\lambda_0}^{\lambda_1} i(\lambda) d\lambda , \qquad (1)$$

where i(λ) is the solar spectral irradiance (power per unit area per unit wavelength). Average reflectances of interest include solar reflectance S (0.3 – 2.5 μ m), UV reflectance U (0.3 – 0.4 μ m), visible reflectance V (0.4 – 0.7 μ m), and NIR reflectance N (0.7 – 2.5 μ m).

It follows from Eq. (1) that the solar reflectance of a surface may be computed as the weighted average of its UV, visible, and NIR reflectances. The aforementioned distribution of solar power (5% UV, 43% visible, and 52% NIR) yields

$$S = 0.05 \ U + 0.43 \ V + 0.52 \ N \ . \tag{2}$$

Strong UV absorption by surface-layer pigments (e.g., titanium dioxide rutile white) or aggregate (e.g., granules) is usually desirable to prevent UV damage to lower components of the roofing product, such as the primer layer in a coated metal system or the asphalt in a granule-surfaced asphalt shingle. High UV reflectance would be even better, but is difficult to achieve with nonmetallic surfaces. Hence, we maximize solar reflectance by establishing high reflectances in the visible and NIR spectra that contain 95% of the incident solar radiation.

Since there is usually more than visible spectral reflectance curve (reflectance versus wavelength in the visible spectrum) that will yield a desired color under a particular illuminant, it is possible to maximize visible reflectance by designing to a color, rather than to a visible spectral reflectance curve. However, this may yield metamerism, in which the color of the coated surface matches that of another surface under one illuminant (e.g., early morning sun) but not another (e.g., noon sun). Maximizing only NIR reflectance avoids this problem.

Creating a coated surface with high NIR reflectance

When appearance and hence visible reflectance are constrained by design, a "cool" surface is one with high NIR reflectance. Consider a substrate (opaque structural material) with a uniformly

pigmented coating. The spectral (wavelength-specific) reflectance of this system depends on the spectral reflectance of the substrate, the thickness of the coating, and on the extent to which light passing through the coating is absorbed (converted to heat) and/or backscattered (reversed in direction) at that wavelength by suspended pigment particles. Reflectance is also influenced by the refractive index of the otherwise-clear coating vehicle. For example, the passage of normally incident collimated light from air (refractive index 1) to a smooth polymer or silicate coating (refractive index 1.5) induces a 4% "first surface" reflection.

Backscattering usually has less effect than does absorption on the reflectance of a substrate with a pigmented coating because some of the light backscattered toward the surface is later backscattered away from the surface. Consider a 25-µm-thick pigmented coating applied to a substrate of reflectance 0.50. Neglecting first surface effects, a nonabsorbing coating with a Kubelka-Munk backscattering coefficient of 5 mm⁻¹ will increase system reflectance by 0.03; of 10 mm⁻¹, by 0.06; of 50 mm⁻¹, by 0.19; of 100 mm⁻¹, by 0.28; and of 200 mm⁻¹, by 0.36. A nonscattering coating with a Kubelka-Munk absorption coefficient of 0.5 mm⁻¹ will decrease system reflectance by 0.01; of 1 mm⁻¹, by 0.02; of 5 mm⁻¹, by 0.11; of 10 mm⁻¹, by 0.20; and of 20 mm⁻¹, by 0.32 (Levinson et al. 2005a). That is, the reflectance decrease induced by absorption can be comparable to the reflectance increase caused by backscattering an order of magnitude larger. Hence, we classify the backscattering by a pigmented coating as "weak" if its backscattering coefficient is less than 10 mm⁻¹, "moderate" if between 10 and 100 mm⁻¹, or "strong" if greater than 100 mm⁻¹. Absorption by a pigmented coating is labeled "weak" if its absorption coefficient is less than 1 mm⁻¹, "moderate" if between 1 and 10 mm⁻¹, or "strong" if greater than 10 mm⁻¹.

A pigmented coating will typically be designed to exhibit strong absorption and/or strong backscattering in the visible spectrum to hide (and thereby color) the substrate. We describe a pigmented coating as "cool" if it has weak NIR absorption, and "hot" if it has strong NIR absorption.

Our survey of the solar spectral radiative properties of common colorants (Levinson et al. 2005b) determined that only a few pigments—e.g., titanium dioxide rutile white, nickel and chrome titanate yellows, aluminum flakes, and mica flakes coated with titanium dioxide—exhibit both strong NIR backscattering and weak NIR absorption when suspended in a vehicle of refractive index 1.5 (Table 1). Some of the nominally cool pigments, such as mixed-metal oxide selective blacks, exhibited both moderate-to-strong backscattering and moderate-to-strong absorption in the NIR. These do not meet our strict requirement (weak NIR absorption) for cool pigments, but may nonetheless be useful for cool applications so long as the coating's NIR reflectance (increased by backscattering, decreased by absorption) is sufficiently high. We note that the manufacturer-reported solar spectral reflectance of a thin (order 25 μ m) pigmented coating on an aluminum substrate (N=0.90) tends to exaggerate the solar spectral reflectance that a similar coating will achieve when applied to a poor NIR reflector, such as a gray-cement concrete tile (N=0.15).

Most cool pigments surveyed exhibited weak-to-moderate backscattering in a vehicle of refractive index 1.5. A thin (but visibly hiding) coating colored with such pigments may be applied directly over a substrate of high NIR reflectance to produce a colored surface with high

NIR reflectance. Some bare roofing materials have NIR reflectances of 0.55 or higher², including wood, treated ZINCALUME[®] steel, treated hot-dipped galvanized (HDG) steel, and natural red clay tile (Table 2). The application of a cool coating to any of these substrates will yield a cool roofing surface with a solar reflectance of at least 0.30. System NIR reflectance will be further increased by NIR backscattering in the cool coating.

Other bare roofing materials, such as gray-cement concrete tiles and gray-rock-surfaced asphalt shingles, have NIR reflectances in the range of 0.10 to 0.15. These substrates with low NIR reflectance can achieve high NIR reflectance via the application of a 25- to 100- μ m thick cool coating pigmented with a moderate-to-strong NIR backscatterer. For example, a smooth, flat, dark gray substrate (N=0.10) with a 25- μ m thick polymer or silicate coating pigmented with titanium dioxide rutile white (backscattering coefficient about 200 mm⁻¹ and absorption coefficient about 0.5 mm⁻¹ at wavelength 1 μ m) achieves an NIR reflectance of about 0.65. N can be increased to over 0.80 by making the coating at least 100- μ m thick (Table 3; Figure 2). A visibly hiding cool topcoat may be applied over this layer to produce an arbitrarily colored system with high NIR reflectance.

Pigmented coatings with moderate-to-high NIR absorption and backscattering tend to yield moderate NIR reflectance over any surface, since the coating is usually NIR opaque. For example, the same gray substrate with a 25-μm thick coating pigmented with an iron oxide red (backscattering coefficient about 100 mm⁻¹ and absorption coefficient about 10 mm⁻¹ at wavelength 1 μm) achieves an NIR reflectance of about 0.40. Increasing the coating thickness to 100 μm (or greater) yields an NIR reflectance limited to about 0.50.

Creating a color-matched cool coated surface

A cool coated surface of a particular color may be created by using a mixture of cool pigments in the topcoat. The topcoat must exhibit strong visible absorption and/or backscattering to hide the substrate (and basecoat, if present). It is crucial to not to adjust the color with pigments that strongly absorb light across the entire solar spectrum, such as carbon black, lampblack, iron oxide black, or copper chromite black. The inclusion of any of these NIR-absorbing pigments in the topcoat or basecoat will tend to make the coated surface hot.

Effect of surface curvature on reflectance

Absorption of multiply reflected light can make the net absorptance of a curved, opaque surface exceed the "local" absorptance that would be observed were the surface flat. We plan to quantify in future research the effect of surface curvature on the reflectances of non-flat roofing surfaces, such as S-shaped clay tiles and granule-covered asphalt shingles.

² Most NIR reflectances presented in this paper are rounded to the nearest 0.05 to avoid unnecessary detail.

Application to Residential Roofing Materials

Metal

A manufacturer of pigmented polyvinylidene fluoride (PVDF) coatings (BASF Industrial Coatings; Colton, CA) provided (a) four types of steel substrate; (b) seven conventionally coated and seven nominally cool-coated ZINCALUME[®] steel coupons; (c) a free film of the polyurethane primer layer used between the metal and the color coat; and (d) 27 free films of PDVF coatings colored with different pigments, one pigment per film.

We measured the solar spectral reflectance of each sample from 0.3 to $2.5 \mu m$ at 5-nm intervals in accordance with ASTM Standard E903 (ASTM 1996), using a PerkinElmer Lambda 900 UV/visible/NIR spectrometer with a Labsphere 150-mm integrating sphere.

The bare and treated samples of HDG steel (steel coated with zinc) and ZINCALUME[®] steel (steel coated with an alloy of 55% aluminum, 43.4% zinc, and 1.6% silicon by mass [Steelscape 2005]) had NIR reflectances ranging from about 0.55 to 0.80 (Table 2; Figure 3). We hypothesize that bare ZINCALUME[®] steel (N=0.80) is appreciably more NIR-reflective than bare HDG steel (N=0.60) because aluminum is more NIR reflective than zinc. Metal treatment—a.k.a. "pretreatment," since it occurs prior to painting—includes cleaning, roughening, and the application of a "conversion layer" to help bind the primer layer to the metal. Treatment decreases the NIR reflectance of ZINCALUME[®] steel by about 0.10 (to N=0.70) and that of HDG steel by about 0.05 (to N=0.55) (Figure 4). This suggests that cool coatings on treated ZINCALUME[®] steel will tend to better reflect NIR radiation than will those on treated HDG steel (N up to 0.15 higher).

Conversion coatings are thin (typically less than 1-µm thick) and contain mostly transparent, non-absorbing inorganic phosphate compounds and polymers of refractive index 1.5 (Roland et al. 1998; Hamacher 1994; Mady and Seidel 1996). Mady and Seidel show Auger depth profiles indicating that two such coatings (one on galvanized steel, and the other on aluminum) are each less than 100 nm thick. One possible reason for the reflectance reduction induced by treatment is that the conversion coating may act as a non-absorbing anti-reflection layer (Born and Wolf, 1999). The reflectance of a coated metal is a function of the ratio of coating thickness to wavelength. Since this ratio approaches the same value (zero) as the film becomes very thin and/or the wavelength grows very large, we expect the effect of the coating on system reflectance to vanish at long wavelengths. We observe in Figure 4 that the reflectance reduction induced by treatment of the two steel samples generally diminishes with increasing wavelength.

The reflectance reduction might also stem from surface roughness induced by etching. For example, a polished aluminum surface, while mirror-like, actually has many submicron grooves produced by the polishing-powder particles, and is less reflective than a smooth aluminum film prepared by evaporation in ultrahigh vacuum (Smith et al. 1985).

The NIR absorptance of the 19- μ m thick sample of primer (polyurethane pigmented with strontium chromate and titanium dioxide) was about 0.03. Since the primer thickness in the 14 coated ZINCALUME[®] samples was only about 5 μ m, any NIR absorption in the coating systems (primer plus PDVF topcoat) occurred almost entirely in the pigmented PDVF layer.

The NIR reflectances of the seven conventionally coated ZINCALUME coupons ranged from about 0.05 to 0.25, while those of the seven cool-coated ZINCALUME coupons ranged from about 0.40 to 0.50 (Figure 5a). This indicates that the nominally cool coatings, while certainly less NIR-absorptive than their color-matched conventional coatings, increased system NIR absorptance by 0.20 to 0.30.

To better understand the nature of the NIR absorptance in the nominally cool pigmented PVDF coatings, and to identify any other pigmented coatings in the manufacturer's product line that might happen to be cool, we characterized the solar spectral radiative properties (reflectance, transmittance, absorptance, backscattering coefficient, and absorption coefficient) of the 27 single-pigment PVDF coatings (Levinson et al. 2005a,b). Many of the nominally cool inorganic pigments exhibited bands of absorption in the NIR induced by the presence of particular elements. For example, pigments that include cobalt were found to have absorption bands centered near wavelength 1.5 µm that cause a 20-µm thick pigmented PVDF coating to absorb about 15 to 40% of NIR radiation. The nominally cool inorganic black pigments containing chromium iron oxide exhibit a gradual, rather than sharp, reduction in absorption from the visible spectrum (where it is desirable) to the NIR spectrum (where it is not), causing a 20-µm thick coating to absorb 35 to 50% of NIR radiation.

A coating pigmented with an organic cool black (perylene black) was found to absorb about 95% of visible radiation, but only about 5% of NIR radiation. When applied in a coating over ZINCALUME[®] steel (*N*=0.70), this weakly scattering pigment can produce a black surface with an NIR reflectance of about 0.65 and a solar reflectance of about 0.35. These values well exceed the NIR (0.35) and solar (0.20) reflectances achieved by ZINCALUME[®] steel with a cool inorganic black PVDF coating.

The total thickness of the coating system (primer plus topcoat) on a metal substrate is typically limited by the need to keep the metal formable without breaking the coating. This tends to make it difficult to significantly increase the NIR reflectance of the system with a basecoat, since the basecoat would have to be quite thin. The additional pass required to apply a basecoat would also increase the cost and reduce the throughput of coil coating processes originally configured for only two layers (primer plus topcoat).

Clay tile

A manufacturer of clay tile roofing (MCA Clay Tile; Corona, CA) supplied 18 clay tile chips (small cut pieces of tile). The NIR reflectance of bare terracotta (natural red) tile was 0.55, while that of glazed tiles ranged from 0.25 (burnt sienna) to 0.75 (white buff). Only two tiles (glazed with burnt sienna and carbon, respectively) had NIR reflectances less than 0.40, and only four tiles had NIR reflectances less than 0.50 (Figure 5b).

Clay tile is typically composed of transparent crystalline particles (size order $10 \mu m$) with anisotropic (directional) refractive indices. Light propagating through the material is scattered when light passes between two differently oriented crystallites that present different indices of refraction. Bare white tile has high reflectance across the NIR spectrum (N=0.85), while bare terracotta tile contains iron oxide (hematite) and therefore exhibits some NIR absorption (Figure 6).

The high NIR reflectance of bare clay tile and the ability to apply thick glazes aid the creation of NIR-reflective glazed clay tiles. It may be possible to modestly increase the NIR reflectance of a glazed tile system by applying and firing a basecoat glaze of white buff before applying and firing the topcoat color glaze. However, it is most important to avoid the use of carbon and other NIR-absorbing pigments in the glaze.

In principle, the solar spectral radiative properties of tile glazes can be characterized in a manner analogous to that used for polymer coatings—i.e., by measurement of the spectral reflectance and transmittance of a glaze applied to a clear substrate, such as quartz, that can withstand firing at 1000°C. However, since the thermal expansion rate of quartz is several times smaller than that of a silicate glaze, a glaze fired on quartz (rather than on clay) will tend to crack. We continue to seek a substrate that is transparent to sunlight, can be fired at high temperature, and thermally expands at a rate comparable to that of the glaze.

We measured the solar spectral reflectances of 20 single-pigment glazes provided at various concentrations on white clay tiles by a manufacturer of tile glazes (Ferro Corporation Frit/Color Division; Los Angeles, CA). These data will be used in our future efforts to improve the NIR reflectances of glazed clay tiles.

Concrete tile

Lawrence Berkeley National Laboratory (LBNL) collaborated with a manufacturer of concretetile coatings (American Rooftile Coatings; Brea, CA) to design 25 prototype acrylic coatings on gray-cement concrete tile chips (*N*=0.15). We divide these into four sets: (A) six conventionally pigmented, 100-μm thick coatings applied directly to tile; (B) a matching set of six nominally cool-pigmented, 100-μm thick coatings also applied directly to tile; (C) the same six nominally cool-pigmented coatings applied at a thickness of 50 μm over a 100-μm thick acrylic white basecoat (*N*=0.85); and (D) seven "experimental," 50- to 150-μm thick topcoats applied over the white basecoat.

The NIR reflectances of the six conventionally coated chips (set A) ranged from 0.05 to 0.55, while those of the nominally cool coatings applied directly to tile (set B) ranged from 0.35 to 0.60. The conventional blue and all the nominally cool coated chips except the black (*N*=0.35) had NIR reflectances exceeding 0.50 (Figure 5c).

The white basecoat used in set C increased the NIR reflectances of the six nominally cool coatings by less than 0.05, suggesting that the original line of cool topcoats was essentially NIR opaque. However, some of the seven experimental coatings in set D were NIR transparent, and show potential to achieve high NIR reflectance when applied over a white basecoat. For example, a 50-µm thick perylene black topcoat applied over the white basecoat achieved an NIR reflectance of 0.55. We note that a 25-µm thick perylene-black PVDF free film with an opaque white background (*N*=0.85) prepared in the course of our pigment characterization activities exhibited an NIR reflectance of 0.85. Hence, it is likely that the NIR reflectance of a gray concrete tile with a white basecoat and a perylene black topcoat can be increased from 0.55 to about 0.85 by reformulating the perylene black topcoat.

Wood shake

Light incident on plant material is backscattered as it passes alternately through cell walls of refractive index 1.4 and intracellular air of refractive index 1 (Knipling 1970). This gives plant material high reflectance at all wavelengths, except in those visible bands where lignin and/or chlorophyll absorb light, and in those NIR bands where water absorbs light (Figure 6). Moderately dark bare wood typically has a visible reflectance of 0.20, an NIR reflectance of about 0.70, and a solar reflectance of about 0.45. Hence, bare wood (and wood shakes treated with an NIR-transmissive fire retardant) can be both cool and dark.

A pigment manufacturer (Ferro Corporation Frit/Color Division; Los Angeles, CA) provided 16 samples of wood (*N*=0.70) with acrylic coatings colored by NIR-scattering inorganic pigments. Each of four pigments (two yellows, one green, and one black) was applied individually to wood in coatings with coverage rates of 150, 125, 100, and 75 ft²/gal, corresponding to mean thicknesses of 270, 235, 405, and 545 μm, respectively. The thickest yellow coatings increased NIR reflectance to 0.80, while the thickest green coating increased NIR reflectance to 0.75. The thickest black coating reduced NIR reflectance to about 0.55.

When a manufacturer of fire retardants for wood products (Galchem Chemical Inc.; Payson, AZ) added metal-oxide pigments to a fire retardant solution that was pressure-applied to wood shakes, the pigments and some of the fire retardant precipitated out of the solution. Further work in this area is needed to investigate the compatibility of pigments with fire retardants.

Asphalt shingle

Over 97% of the surface of a typical asphalt-soaked fiberglass roofing shingle is covered with a layer of crushed rocks, or "granules," that are about 0.5 to 2 mm in diameter (Akbari et al. 2005a). Hence, the NIR reflectance of an asphalt shingle is determined by that of its granule layer. The NIR reflectance of the granule layer is in turn is limited by (a) the low NIR reflectance of typical gray-rock granules (about 0.10 - 0.15); (b) the low mean thickness of a typical granule coating (about 5 to 10 μ m); and (c) weak to moderate NIR backscattering by most pigments.

The simplest way to increase the NIR reflectance of individual granules is to use a naturally white (or otherwise light-colored) aggregate. However, some light-colored rocks such as quartz transmit UV light, and would fail to shield the asphalt from UV radiation in sunlight. If a UV-opaque, NIR-reflective aggregate is not available, an NIR-reflective basecoat pigmented with a titanate white, a titanate yellow, titanium-dioxide coated mica flakes, or aluminum flakes can be applied to an NIR-absorbing aggregate to produce an NIR-reflective granule. For example, a 5-µm thick coating of refractive index 1.5 that is pigmented with titanium dioxide white can increase the NIR reflectance of a smooth, dark gray surface (*N*=0.10) to 0.35. A 10-µm thick coating will increase NIR reflectance to 0.50; a 25-µm thick coating, to 0.65 (Table 3; Figure 2). A cool, visibly hiding topcoat can provide color and, optionally, additional NIR backscattering.

The thickness of the coating applied to a granule is limited by the coating process, in which granules are preheated in a tumbler; transferred hot to a rotary mixer for application of the wet pigmented coating (pigments in sodium silicate, hydrated kaolin clay, and water); and then fired in a rotary kiln. If the volume ratio of liquid coating to granules is too high, the granules will

tend to fuse together. Multiple passes increase the total coating thickness, but reduce system throughput and increase cost.

LBNL collaborated with a manufacturer of roofing granules (ISP Mineral Products; Hagerstown, MD) to develop approximately 90 prototype shingle coupons (40 cm²) and 10 prototype shingle boards (0.5 m²). The shingles were surfaced with (a) bare rock granules; (b) granules with thin or thick white coatings; (c) granules with a thin aluminum coating; (d) granules with cool-pigment topcoats over bare rock, rock with an aluminum basecoat, rock with a thin white basecoat, or rock with a thick white basecoat; (e) salt-and-pepper blends of bright-white granules (rock with a thick white coating) and some of the granules described in (d); and (f) blends of granules of varying sizes.

The NIR reflectance of a shingle surface covered with bare granules was about 0.10. Adding a topcoat colored with a black, brown, green, or blue cool pigment increased NIR reflectance to 0.10-0.30. A thin (about 15 µm) aluminum basecoat (granulated surface N=0.35) increased NIR reflectances of the colored granules to about 0.25; a thin (about 15 µm) white basecoat (granulated surface N=0.25), to 0.25 – 0.35; and a thick (about 25 µm) white basecoat (granulated surface N=0.45), to 0.30 – 0.45 (Figure 5d).

Adding a basecoat tended to increase both visible and NIR reflectances unless the topcoat was opaque to visible light. For example, a coating colored with a cool inorganic brown pigment produced a shingle with V=0.15 and N=0.28 when applied over a bare granule. Adding a thin white basecoat yielded V=0.22 and N=0.35; adding a thick white basecoat yielded V=0.26 and V=0.43. The increases in the visible reflectance of this shingle were comparable to its increases in NIR reflectance—that is, the shingle became lighter in color. However, the shingle was not spectrally gray, in the sense that its NIR reflectance well exceeded its visible reflectance.

Figure 7 illustrates the development of cool black asphalt shingle colored with an inorganic cool black pigment. The granules on a conventional shingle are pigmented with carbon—a hot black colorant with strong absorption across the entire solar spectrum—and have no basecoat. In prototype 1, the carbon is replaced by a cool inorganic black, increasing NIR reflectance from 0.05 to 0.19 and solar reflectance from 0.04 to 0.12. Prototype 2 adds a thin white basecoat below the cool inorganic black topcoat, increasing *N* to 0.26 and *S* to 0.16. Prototype 3 replaces the thin white basecoat with a thick white basecoat, increasing *N* to 0.30 and *S* to 0.18. The top curve ("performance limit") corresponds to a smooth, 25-μm-thick PDVF film (*N*=0.43, *S*=0.25) pigmented with this cool black and backed by an opaque white (*N*=0.85). Roughness and limits to the thicknesses of the basecoat and topcoat are expected to make the reflectance of any granule-surfaced shingle pigmented with this cool inorganic black less than that achieved by the white-backed smooth film.

The prototype salt-and-pepper blends replaced thinly coated white granules with thickly coated white granules; some also replaced standard color granules with cool color granules. Unsurprisingly, using whiter granules increased both visible and NIR reflectances, resulting in a lighter-colored shingle with higher solar reflectance.

Blending various sizes of granules to make the granule layer smoother did not noticeably increase reflectance.

It may be possible to increase the NIR reflectance of granule-surfaced asphalt shingles by applying the pigmented coating to the shingle after bare granules have been pressed into the asphalt, then baking the granulated surface with radiant heat. This approach would coat only the exposed faces of the granules and might mitigate thickness limits associated with the tumble-coating/kiln-drying processes.

We note that several other roofing manufacturers, including CertainTeed (Valley Forge, PA), 3M Industrial Minerals (St. Paul, MN), and Elk Corporation (Dallas, TX) are engaged in analogous efforts to produce cool nonwhite asphalt shingles. The CertainTeed process applies cool pigments to both the roofing granules and the asphalt substrate (Shiao et al. 2005a,b), while the 3M process applies a solar-reflective basecoat and a cool colored topcoat to the granules (Gross and Graham 2005). The Elk process has not been disclosed.

Summary

Surfaces with high thermal emittance (i.e., nonmetals, and most polymer-coated metals) stay cool in the sun when they have high solar reflectance. When strong UV absorptance is required to shield a substrate and visible spectral reflectance is fixed to yield a particular color, maximizing NIR reflectance is equivalent to maximizing solar reflectance. The NIR reflectance of a substrate with a pigmented coating generally depends on NIR absorption and backscattering in the pigmented coating, and on the NIR reflectance of the uncoated substrate. Cool coatings should exhibit low NIR absorption. However, some NIR absorption may be acceptable in a coating that also exhibits strong NIR backscattering. A substrate with high NIR reflectance (e.g., metal, clay tile, or wood) can be colored with any cool coating, while a substrate with low NIR reflectance (such as gray-cement concrete tile or gray aggregate) requires significant NIR backscattering in either the cool topcoat or a cool basecoat.

Coated metal and glazed clay tile roofing products achieved NIR reflectances of up to 0.50 and 0.75, respectively, using only a cool topcoat. Topcoats colored with NIR-transparent organic pigments could yield coated metal systems with NIR reflectances as high as 0.65. Gray-cement concrete tiles have low NIR reflectance, but achieved NIR reflectances as high as 0.60 when thickly coated with NIR-scattering pigments. Coated gray-cement concrete tiles with NIR reflectances as high as 0.85 could be obtained by overlaying a titanium-dioxide basecoat with a topcoat colored by NIR-transparent organic pigments. Granule-surfaced asphalt shingles achieved NIR reflectances as high as 0.45 when a cool color topcoat was applied to granules with a thick white basecoat. Bare wood has an NIR reflectance of about 0.70; the application of certain pigments (e.g., metal oxides) may remove fire retardants from wood roofing products, and is not recommended.

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Table 1. Classification of cool pigments (those with low NIR absorption) according to strength of NIR backscattering in a vehicle of refractive index 1.5.

| Strong NIR backscattering | Moderate NIR backscattering | Weak NIR backscattering |
|---|---|--|
| (coefficient $> 100 \text{ mm}^{-1} \text{ at } 1 \mu\text{m}$) | (coefficient 10 - 100 mm ⁻¹ at 1 μm) | (coefficient $< 10 \text{ mm}^{-1} \text{ at } 1 \mu\text{m}$) |
| chrome titanate yellow | cadmium orange, yellow | cobalt aluminate blue |
| chromium iron oxide black* | cobalt chromite blue, green* | diarylide yellow |
| nickel titanate yellow | cobalt titanate green | dioxazine purple |
| titanium dioxide (rutile) on | iron titanium brown spinel | Hansa yellow |
| mica flakes (interference | modified chromium oxide | perylene black |
| colors) | green | phthalocyanine blue, green |
| titanium dioxide rutile white | monastral red | quinacridone red |
| | red, brown iron oxides | ultramarine blue |

^{*} These pigments exhibit moderate absorption in the NIR and hence are not strictly cool, but have sufficient NIR backscattering to be useful in cool coatings.

Table 2. Typical NIR reflectances (rounded to nearest 0.05) of various uncoated substrates.

| Uncoated substrate | NIR reflectance |
|--------------------------------------|-----------------|
| fresh asphalt | 0.05 |
| layer of gray-rock granules | 0.10 |
| gray-cement concrete tile | 0.15 |
| treated* hot-dipped galvanized steel | 0.55 |
| bare hot-dipped galvanized steel | 0.60 |
| natural red clay tile | 0.70 |
| wood | 0.70 |
| treated* ZINCALUME® steel | 0.70 |
| bare ZINCALUME® steel | 0.80 |
| white ceramic tile | 0.85 |
| aluminum foil | 0.90 |

^{*}cleaned and coated with a very thin (about 1 μm) "conversion layer" that helps the primer layer adhere to the metal

Table 3. NIR reflectances (rounded to nearest 0.05) of white and aluminum-flake acoatings on a smooth, dark gray substrate (N=0.10).

| Coating | NIR reflectance of coated substrate |
|-----------------------|-------------------------------------|
| none | 0.10 |
| white, 5 μm | 0.35 |
| white, 10 μm | 0.50 |
| white, 25 μm | 0.65 |
| white, 50 μm | 0.75 |
| aluminum flake, 25 µm | 0.80 |
| white, 100 μm | 0.80 |
| white, 200 µm | 0.85 |

^{*} Polyvinylidene fluoride (PVDF) pigmented with titanium dioxide rutile (mean particle size 0.25 µm) at 15% volume concentration. Comparison of measured and computed values of the NIR reflectance of coatings pigmented with titanium dioxide white suggests that these computed values may be slightly (as much as 0.05) too low.

^{**} Silicone pigmented with aluminum flakes. It may be possible to achieve a comparable NIR reflectance with a thinner coating because the aluminum flake coating was NIR opaque at a thickness of about 25 µm.

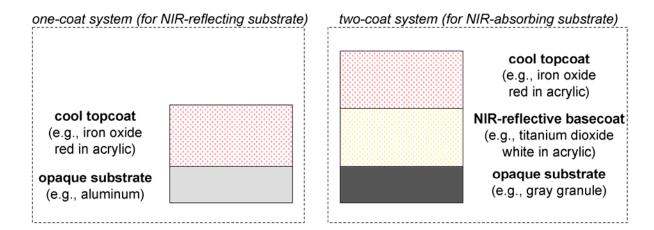


Figure 1. Schematics of one-coat (substrate + topcoat) and two-coat (substrate + basecoat + topcoat) systems. The one-coat system can also be applied over an NIR-absorbing substrate if the topcoat has at least moderate NIR backscattering and is sufficiently thick.

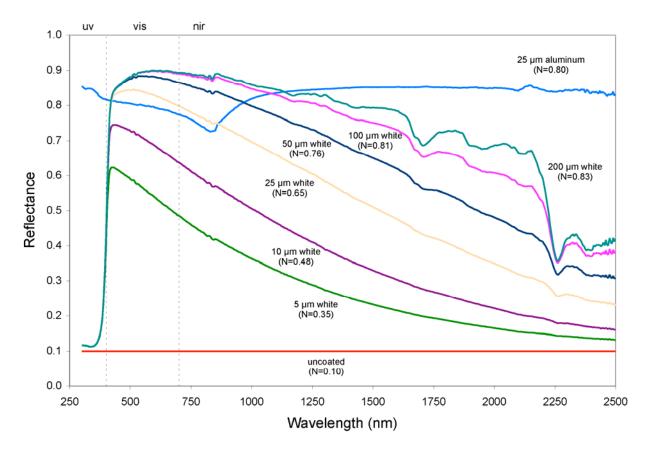


Figure 2. Solar spectral reflectances and NIR reflectances (N) of a smooth, dark gray substrate (reflectance 0.10 at all solar wavelengths) with (a) 5, 10, 25, 50, 100, and 200-μm thick PVDF coatings pigmented with titanium dioxide rutile white at 15% pigment volume concentration; and (b) a 25-μm thick silicone coating pigmented with aluminum flakes (pigment volume concentration unknown). The spectral reflectances of the white-coated surfaces were estimated from absorption and backscattering coefficients computed by Levinson et al. (2005a), while that of the aluminum-flake coated surface was measured.

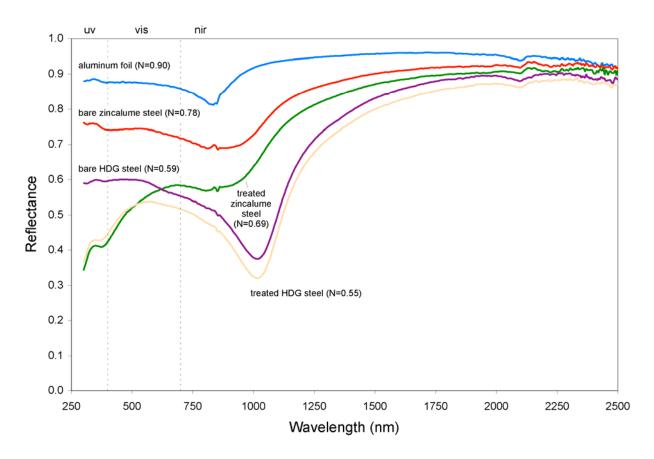


Figure 3. Solar spectral reflectances and NIR reflectances (N) of five uncoated, metallic substrates: aluminum foil, bare and treated ZINCALUME[®] steels, and bare and treated hot-dipped galvanized (HDG) steels.

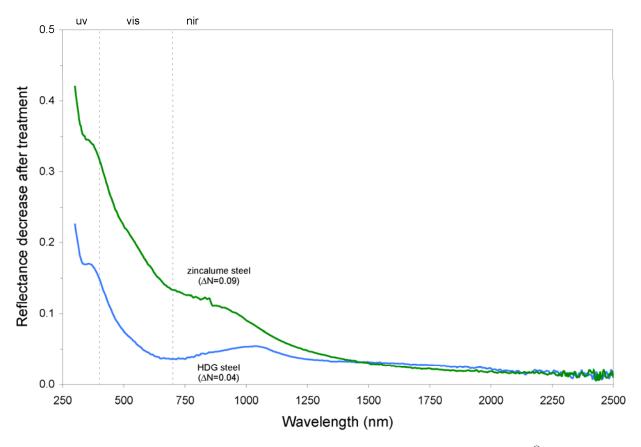


Figure 4. Decreases in spectral reflectance induced by treating ZINCALUME[®] and hot-dipped galvanized (HDG) steels. Shown also is the decrease ΔN in NIR reflectance.

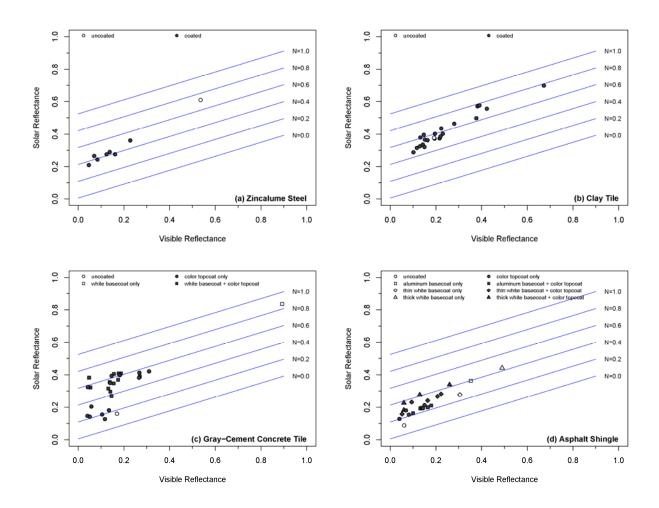


Figure 5. Solar vs. visible reflectances of uncoated and cool-coated samples of (a) treated ZINCALUME® steel, (b) clay tile, (c) gray-cement concrete tile, and (d) granule-surfaced asphalt shingles. Lines of constant NIR-reflectance N assume a UV reflectance of 0.1.

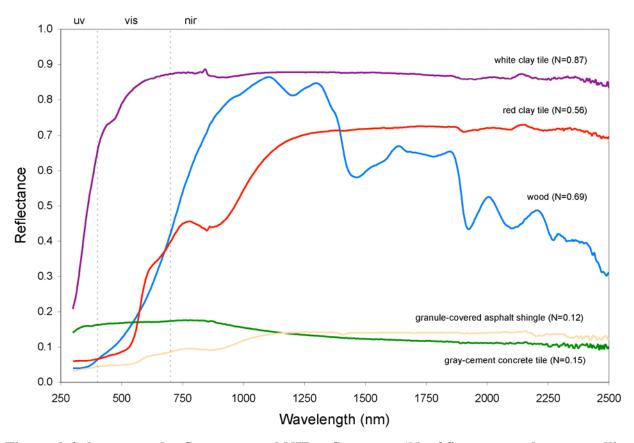


Figure 6. Solar spectral reflectances and NIR reflectances (N) of five uncoated, nonmetallic substrates: white clay tile, red clay tile, wood, granule-covered asphalt shingle, and gray-cement concrete tile.

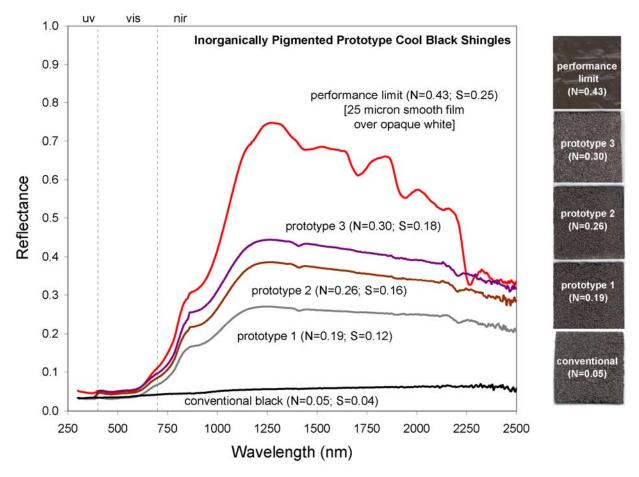


Figure 7. Development of an inorganically pigmented cool black asphalt shingle (N=near-infrared reflectance; S=solar reflectance).